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Plasmon resonance enhanced mid-infrared generation by graphene on gold gratings through difference frequency mixing

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ABSTRACT

Graphene has been demonstrated to have extraordinary large second order nonlinear susceptibility that can be applied in generating mid-infrared (MIR) and terahertz waves through the difference frequency process. In this study, we exploit the highly localized electric fields caused by plasmon resonances to increase the nonlinear response from graphene. The proposed structure contains a graphene sheet on a gold grating substrate that sustains both surface plasmons at the near-infrared on the gold surface and plasmons at the MIR on the graphene surface. Based on finite difference time domain (FDTD) numerical simulations, more than 3 orders of magnitude improvement of the MIR generation efficiency is obtained by placing graphene sheets on a gold grating substrate under resonance conditions instead of placing them on a flat substrate. With the same gold grating substrate, MIR waves tunable from 30 to 55 THz are generated by tuning the gate voltage of the graphene sheet.

1. Introduction

Ever since its first isolation in 2004, graphene (a single layer of carbon packed in a honeycomb lattice) has received enormous interest for its remarkable electrical, optical and physical properties [1–3]. Aside from these interesting properties, the extraordinary large nonlinear optical response has drawn much attention in recent years. Third order nonlinear processes, such as optical Kerr effect, third harmonic generation and four wave mixing, have already been experimentally verified by single or few layer graphene thin films [4–6]. Second order nonlinear processes such as difference frequency generation (DFG) are usually assumed to be forbidden in an isotropic medium, but possible in graphene because of the distinctively non-local, spatial character of the interaction. The second order nonlinear susceptibility of graphene of $\chi^{(2)} \sim 3 \times 10^{-7} m/\mathrm{V}$ is experimentally measured by taking its thickness of 0.3 nm into account [7], which is approximately three orders of magnitude larger than in GaAs. Even larger values of $\chi^{(2)}$ are predicted in theoretical studies [8–10]. As an example of the applications taking advantage of this large nonlinearity, graphene surface plasmons at mid-infrared (MIR) and terahertz (THz) wavelengths have been coherently excited using two visible frequency free space beams via DFG [7].

Despite of the surprisingly large intrinsic nonlinear susceptibility of graphene, nonlinear optical effects can be further enhanced by the presence of highly confined plasmons. When plasmon resonance takes place, the local electric field is dramatically enhanced, which results in an improved nonlinear conversion efficiency. For example, improved efficiency of third harmonic generation due to field enhancement caused by plasmonic resonances from graphene on a dielectric grating substrate has been reported, recently [11]. However, in previous studies such as references [7,8], only field enhancement at the MIR or THz wavelength is utilized, the excitation waves at visible wavelengths have no enhancements. As we all know, noble metals, such as gold, can support surface plasmon polaritons (SPPs) in the visible and near-infrared (NIR) wavelengths [12–14]. It may be possible to combine the NIR SPPs of noble metals and the MIR SPPs of graphene in applications such as MIR generation.

Here, we introduce the model of a graphene sheet on a gold grating substrate to obtain enhanced nonlinear conversion of MIR waves through DFG by exploiting both the field enhancements at the incident NIR wavelengths and the generated MIR wavelength. To bridge the large mismatch between the SPP wave vectors and the free space beams, the NIR SPPs on gold surface are excited by the fundamental mode of the grating, while the MIR SPPs on graphene surface is excited by the high order mode of the grating. Certified by numerical simulations based on the finite difference time domain (FDTD) method, more than 3 orders of magnitude improvement of the MIR generation efficiency is obtained by placing graphene on a gold grating substrate under resonance conditions instead of placing them on a flat substrate.

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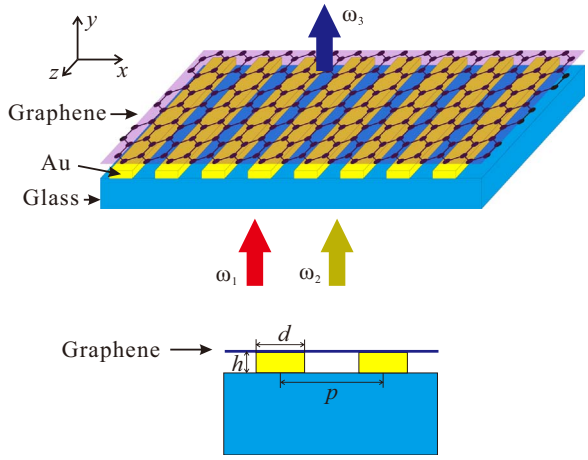

Fig. 1. Schematic of the model designed to generate enhanced MIR waves from the graphene sheet on a gold grating substrate. Two input beams at frequencies of ω_1 and ω_2 interact with each other in the graphene sheet to generate the difference frequency beam at $\omega_3 = \omega_1 - \omega_2$. The gold grating has a period of p. Each gold strip has a height of h and a width of d.

2. Model

The proposed structure to generate plasmon resonance enhanced MIR waves is depicted in Fig. 1. As can be seen, a graphene layer is deposited on a gold grating substrate with a period of p, which is further held by a glass substrate. Each gold strip has a height of h and a width of d. In the DFG process, two intense beams at frequencies of ω_1 and ω_2 illuminate the sample normally and interact with each other in the graphene sheet to generate the difference frequency beam at $\omega_3 = \omega_1 - \omega_2$. This process can be expressed by a nonlinear polarization according to [15].

$$\mathbf{P}^{(2)} = 2\varepsilon_0 \chi^{(2)} \boldsymbol{E}_1 \boldsymbol{E}_2^* \tag{1}$$

where ε_0 is the permittivity of free space, $\chi^{(2)}$ is the second order nonlinear susceptibility of graphene, and $\boldsymbol{E}_i$ are the electric field vectors associated with the incident beams at frequencies of ω_i.

Without loss of generality, the wavelengths of the pump and the signal beams are chosen to be λ_1=800nm and λ_2=895.52nm, respectively. Thus, the generated difference frequency beam has a wavelength of λ_3=7.5μm, according to the energy conservation law of the DFG process. These wavelengths of the incident beams are experimentally accessible with a femtosecond Ti: sapphire laser combined with an optical parametric oscillator. In our designed structure, the pump and the signal beams at NIR experience plasmon resonances on the gold surface, while the DFG beam at MIR experience plasmon resonance on the graphene surface. As a result, the local electric fields of all three beams (E_i) are enhanced, which further leads to a dramatic enhancement of the DFG process. As a comparison, we denote the electric fields in the model of a graphene layer placed on a flat glass substrate as $E_{flat,i}$. Thus, the local enhancement factor for the electric field can be defined as $\eta_i = |E_i/E_{flat,i}|$ for the beam at frequency of ω_i. Therefore, the local enhancement factor of the DFG process can be written as

$$\mathrm{EF} = (\eta_1 \eta_2 \eta_3)^2 \tag{2}$$

The local enhancement factor defined here is in fact the ratio $\mathrm{EF} = P_{grating}/P_{flat}$, where $P_{grating}$ and P_{flat} are the powers collected at ω_3 when the graphene sheet on a gold grating substrate and graphene on a flat substrate, respectively, are illuminated by two beams at frequencies ω_1 and ω_2, considering only an infinitesimal volume of the graphene sheet. The overall DFG generation power can be calculated by summing the power generated by each volume element of the graphene sheet, as obtained in the FDTD simulations. Thus the enhancement factor of the electric fields (EF) is proportional to the enhancement factor of the generation efficiency.

3. Simulation parameters

To analyze the plasmon-assisted DFG process, we perform numerical simulations with the finite-difference time-domain (FDTD) method using Lumerical FDTD solution. Since the proposed structure is invariant in the z direction, the simulations are performed in the two-dimensional case without loss of accuracy. Only one period of the grating is needed in the simulation region by enforcing periodic boundary conditions in the x direction. Perfectly matched layer (PML) boundary conditions are employed in the y direction.

In the simulations, the graphene sheet is modeled as a thin layer with thickness $t = 0.5\mathrm{nm}$. The linear permittivity of graphene is uniaxial anisotropic, which can be described by a diagonal tensor. Here, the in-plane component of the linear permittivity is set as $\varepsilon_{xx} = \varepsilon_r + i\sigma/(\varepsilon_0 \omega t)$, where ε_r=2.5 is the background relative permittivity and σ is the conductivity of graphene. The surface-normal component is set as $\varepsilon_{yy} = \varepsilon_r$. At the MIR frequencies that SPPs on the graphene occur, the conductivity of graphene is dominated by the intraband transition, which can be approximated by a Drude-like expression as [16,17].

$$\sigma(\omega) = \frac{ie^2\mu_c}{\pi\hbar^2(\omega + i\tau^{-1})} \tag{3}$$

where e is the electron charge, μ_c is the Fermi energy, $\hbar$ is the reduced Planck's constant, τ is the intrinsic relaxation time and assumed to follow the relation $\tau = \mu\mu_c/(ev_f^2)$. Here, v_f=c/300 is the Fermi velocity (c is the speed of light) and $\mu = 1m^2V^{-1}s^{-1}$ is the carrier mobility. The second order nonlinear susceptibility of graphene $\chi^{(2)} \sim 1.8 \times 10^{-7} m/\mathrm{V}$ is applied here, according to the experimental measurement [7]. An intriguing property of graphene plasmonics is its wide tunability. In our proposed model, top gate based schemes such as the scheme in Ref. [18] can be used to tune the graphene Fermi level. In the following simulations, graphene plasmons for Fermi levels from 0.3 to 1 eV are studied, which can be realized by adjusting the applied gate voltage experimentally. Gold simulated in our model is defined as “Au (Gold) - Palik” from the FDTD default material database. The dispersion relation of gold is fitted according to material data in Ref. [19].

Since there is a large dimensional difference between the thickness of the graphene (0.5 nm) and the grating periods (several hundred nanometers), we use non-uniform mesh in the FDTD simulations. The mesh size inside the graphene layer is 0.1 nm, and the mesh size gradually increases outside the graphene layer.

4. 4, Characterization of plasmon resonances

4.1. Plasmon resonance on the gold surface

As can been seen from Eq. (2), field enhancements of all the three beams participating in the DFG process have contributions to the total enhancement of the MIR generation. So, in order to obtain the highest conversion efficiency, we need to find the conditions to excite SPPs on the gold surface and SPPs on the graphene surface, simultaneously. In our method, we firstly tune the grating period to satisfy the excitation condition of the gold SPPs and obtain graphene SPPs by tuning the graphene Fermi level, secondly.

SPPs supported by noble metals have been extensively studied in the past years for their promising applications in nanophotonics. For a gold-dielectric interface, the SPP wave vector obeys the dispersion relation

$$k_{Auspp} = \frac{\omega}{c}\sqrt{\frac{\varepsilon_{Au}(\omega)\varepsilon_d}{\varepsilon_{Au}(\omega) + \varepsilon_d}} \tag{4}$$

where $\varepsilon_{Au}(\omega)$ and ε_d are the permittivity of gold and the dielectric material, respectively. To match this wave vector, schemes like Kretschmann-Raether configuration and the Otto configuration, or